Superconductivity in β-pyrochlore superconductor KOs_2O_6: treatment within strong-coupling Eliashberg theory

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We study the influence of the rattling phonons on superconductivity in β-pyrochlore KOs_2O_6 compound based on the strong-coupling Eliashberg approach. In particular, analyzing the specific heat data we find that the rattling phonon frequency changes discontinuously at the critical temperature of the first order phase transition. Solving the strong-coupling Eliashberg equations with temperature dependent $\alpha^2F(\omega)$, we investigate the consequence of this first order phase transition for the anomalous temperature dependence of the superconducting gap. We discuss our results in context of the recent experimental data.

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The superconductivity in the family of the β-pyrochlore oxides KOs_2O_6, $RbOs_2O_6$, and $CsOs_2O_6$ exhibits many exotic properties[1,2,3,4,5,6]. Among them, KOs_2O_6 with superconducting transition temperature $T_c = 9.6$K shows the most anomalous behavior. For example, the electrical resistivity demonstrates strong concave T dependence down to T_c[1], in contrast to the normal T^2 behavior in Rb and Cs compounds[2,3,4,5,6]. The specific heat measurements have found an existence of low frequency Einstein modes and that the T-linear coefficient of the specific heat $\gamma = 70mJ/K^2 \cdot mol$, (see Refs. 7,8), is strongly enhanced over the value obtained from band structure calculations[9,10]. The band structure calculations have indicated that these anomalies may be due to highly anharmonic low frequency rattling motion of the alkali-ions inside an oversized cage formed by the Os and O ions[9]. Furthermore, this is consistent with the x-ray observation of anomalously large atomic displacement for the K ions[11], and the low frequency phonon structures seen in photoemission spectra[12]. Moreover, recent NMR data[13] have indicated that the relaxation at the K sites is entirely caused by fluctuations of the electric field gradient ascribed to the highly anharmonic low frequency dispersionless oscillation (rattling) of K ions in the cage. Recently, Dahm and Ueda have developed the phenomenological model to describe the influence of the anharmonic phonons on the NMR relaxation and the electrical resistivity[14].

Various experiments have been carried out in KOs_2O_6 to elucidate the mechanism of the superconductivity. Most importantly, thermal conductivity[15] and laser photoemission spectroscopy(PES)[12] measurements have revealed almost isotropic s-wave superconducting gap. This means that the Cooper-pairing can be ascribed to phonons. Furthermore, an absence of the coherence peak in the K-NMR relaxation rate can be explained due to strongly overdamped phonons[13]. At the same time, a large value of the electron-phonon coupling constant $\lambda_{ep} \approx 2.38$, the presence of the low-energy rattling phonon modes and their influence on the electronic properties, and finally, the first-order structural phase transition at $T_p < T_c$[8] makes an investigation of superconductivity in KOs_2O_6 very interesting, in particular, from a point of view of studying further aspects of the strong coupling Eliashberg theory in presence of coupling between conduction electrons and rattling phonons. Previously, an attempt to consider the effect of the temperature-independent rattling phonons has been considered in Ref.16 in application to $CsOs_2O_6$.

In this Brief Report, we analyze the superconductivity in KOs_2O_6 using the standard Eliashberg formalism supplemented by a quasiharmonic treatment of the rattling mode and its renormalization by the coupling with conduction electrons. We find that the phonon spectrum can be modeled by the two Lorentzians peaked at the energies of the two Einstein modes, ω_{E1} and ω_{E2} representing the lowest energy rattling phonons. An additional contribution arises from the Debye frequency at ω_D. To explain the superconducting transition temperature in KOs_2O_6 we have employed the mean-field analysis of the temperature dependence of the lowest Einstein mode proposed previously[14]. We show that its energy should decrease with temperatures down to $T_p < T_c$ and then jump to a higher frequency indicating the first order structural phase transition. Solving the non-linear Eliashberg equations below T_c with temperature dependent electron-phonon coupling function, $\alpha^2F(\omega)$ we compare our results with various experiments.

On the real frequency axis the finite temperature Eliashberg equations for the superconducting gap $\Delta(\omega, T)$ and the renormalization function $Z(\omega, T)$ are

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given by[17]:

$$\Delta(\omega,T) = \frac{1}{Z(\omega,T)}\int_0^\infty d\omega' \mathrm{Re}\left\{\frac{\Delta(\omega',T)}{\sqrt{\omega'^2-\Delta^2(\omega',T)}}\right\} \times \left[K_+(\omega,\omega',T)-\mu^*\tanh\left(\frac{\beta\omega'}{2}\right)\right] \quad , \quad (1)$$

$$\omega(1-Z(\omega,T)) = \int_0^\infty d\omega' \mathrm{Re}\left\{\frac{\omega'}{\sqrt{\omega'^2-\Delta^2(\omega',T)}}\right\} \times K_-(\omega,\omega',T) \quad , \quad (2)$$

where

$$K_\pm(\omega,\omega',T) = \int_0^\infty d\Omega\, \alpha^2F(\Omega)\left[\frac{f(-\omega')+n(\Omega)}{\omega'+\omega+\Omega} \pm \frac{f(-\omega')+n(\Omega)}{\omega'-\omega+\Omega} \mp \frac{f(\omega')+n(\Omega)}{-\omega'+\omega+\Omega} - \frac{f(\omega')+n(\Omega)}{-\omega'-\omega+\Omega}\right], \quad (3)$$

and

$$\alpha^2F(\Omega) = \alpha^2_{E1}F_{E1}(\Omega)+\alpha^2_{E2}F_{E2}(\Omega)+\alpha^2_PF_P(\Omega) \quad , \quad (4)$$

is the generalized electron-phonon coupling function averaged over the Fermi surface. We assume that there are two contributions to the Eliashberg function. The second is arising from the usual dispersive acoustic phonons limited by the Debye energy $T_D = 325$ K. The first contribution is due to local low energy rattling phonons whose energies we denote by ω_{E1} and ω_{E2}. The latter requires a special consideration.

It has been shown recently that the low-energy phonons ascribed to the heavy rattling of the K ions confined in an oversized cage made of OsO_6 octahedra are responsible for the unusual scattering processes in KOs_2O_6[8]. In particular, it has been assumed that contrary to the $RbOs_2O_6$ and $CsOs_2O_6$ cases where a single Einstein mode due to rattling motion is enough, there exits two modes at $\hbar\omega_{E1} = 22$K, and $\hbar\omega_{E2} = 61$K in KOs_2O_6 corresponding to the first two excited energy levels of the corresponding anharmonic potential. Simultaneously, the mean-field description of the local alkali-ion anharmonic motion has been developed in Ref.[14]. Starting from the standard anharmonic Hamiltonian

$$H = \frac{p^2}{2M}+\frac{1}{2}ax^2+\frac{1}{4}bx^4 \quad , \quad (5)$$

where x, p, and M are the spatial coordinate, momentum, and mass of the alkali ion, respectively and a, b are constants with $b > 0$ one finds that according to *ab initio* calculations[9] for KOs_2O_6, the quadratic term becomes negative, *i.e.* $a < 0$, that results in a double well potential. We note that in reality the double wells are not in the plane[9]. Treating the Hamiltonian (5) in the self-consistent quasiharmonic approximation Dahm and Ueda have found that the oscillation of Potassium ions can be described by an effective harmonic oscillation with effective low-energy frequency that now depends on temperature[14]. In particular, it decreases monotonically upon decreasing temperature[18]. At low enough temperatures ($T \ll \omega_0$), this frequency becomes nearly temperature independent. Obviously this model requires certain modification when applied to KOs_2O_6. In particular, although this effective model works well at high temperatures, it fails to explain the occurrence of the first order phase transitions at temperature $T_p < T_c$ and the occurrence of the second Einstein frequency at higher energy.

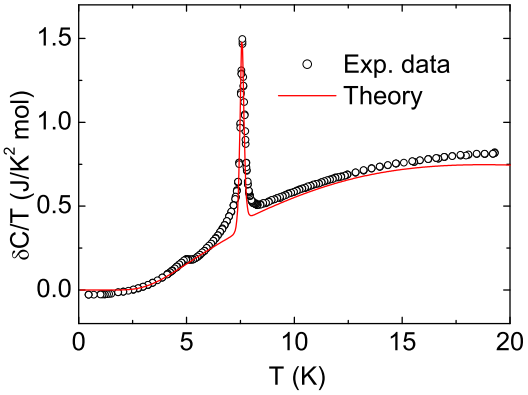

FIG. 1: (color online) Theoretical fit to the experimental specific heat data from Ref. 8 using $\hbar\omega_{E2} = 59$K and $\hbar\omega_{E1}(T = T_c) = 27.4$K. Assuming the jump of the characteristic rattling phonon frequency at T_p from $\hbar\omega_{E1}$=27.4K to $\hbar\omega_{E1}$=30K we find the correspondent increase in the specific heat curve in qualitative agreement with experiment data.

Therefore, modeling the contribution of the rattling K ions to the Eliashberg function we adopt two Einstein like modes centered at ω_{E1} and ω_{E2}. Furthermore, to take into account the upper lying energy levels of the shallow potential we use the self-consistent quasiharmonic approximation for the lowest mode, ω_{E1} that results in its temperature dependence. Most importantly, before entering the temperature independent regime the effective rattling phonon frequency shows a discontinuous jump towards higher frequency that will result in the specific heat anomaly at T_p. The increase of the ω_{E1} at T_p is in accordance with recent experiments showing no structural changes below the phase transition[8,13,20]. Indeed, below T_p the K ions seem to be stabilized in their equilibrium positions which corresponds to the increase of ω_{E1}.

To find the exact shift of the lowest frequency at T_p, and also the approximate positions of the rattling mode frequencies we made a simulation of the specific heat data around T_p following the model proposed by Ref. 8 with temperature dependent ω_{E1}. The contribution from the two frequencies representing the rattling modes is given by[8]

$$C = aC_{E1}+(1-a)C_{E2} \quad (6)$$

where $a = 0.24$ and ω_{E_i} with $i = 1, 2$ are the corre-

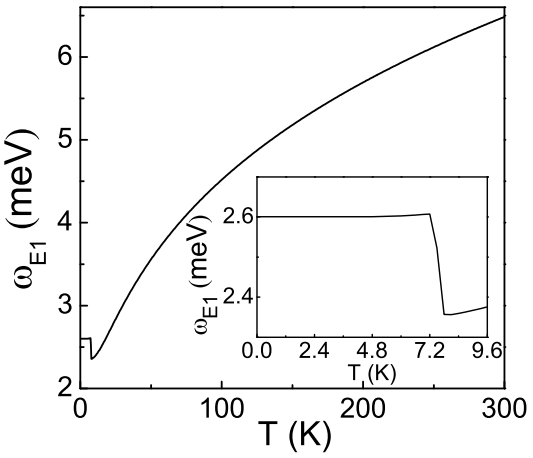

FIG. 2: Calculated temperature dependence of the lower frequency, ω_{E1} of the rattling phonon as obtained using the quasi-harmonic approximation[14]. The parameter of the model $b\hbar/M^2\omega_0^3(T = 300K) = 0.25$ have been fixed by assuming the room temperature position of the rattling phonon frequency around 6 meV as found by neutron scattering experiments[22]. At T_p the effective frequency demonstrates the discontinuous jump to lower energy (as shown in the inset) which position is determined by the absolute value of the specific heat jump shown in Fig.1.

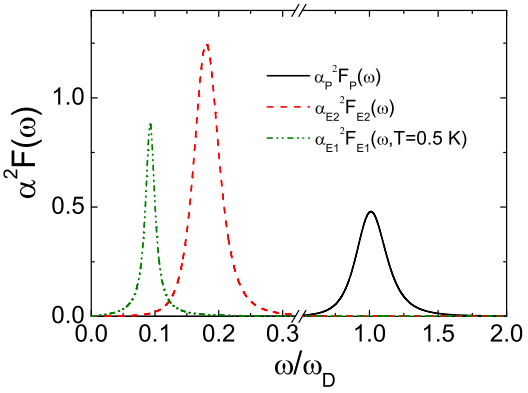

FIG. 3: (color online) Calculated Eliashberg function, Eq.(4), for KOs_2O_6 for T=0.4K. The solid curve denotes the usual phononic contribution centered around Ω_D and the (red) dashed curve refer to the upper rattling phonons frequency centered around ω_{E2}. Here, we employ $\alpha^2F_i(\omega) = \frac{c_i\omega_i\Gamma_i^3}{\pi\left((\omega-\omega_i)^2+\Gamma_i^2\right)^2}$ where $i = E2, p$. We further use $\Gamma_i = \omega_i/5$, $c_{E2} = 0.767$, and $c_p = 0.3$. The (green) dashed-dotted curve refers to the lower energy rattling mode contribution to $\alpha^2F(\omega)$ with parameters as described in the text.

sponding Einstein phonons. As mentioned above, for the sake of simplicity we assume that the higher mode is temperature independent and the lower one follows the mean-field temperature dependence that originates from integrating out the upper-lying energy levels with temperature. Generally, for the contribution of temperature dependent Einstein frequency one finds

$$C_E = 3R\left(\frac{\hbar\omega_E}{k_BT}\right)^2 \frac{\exp\left(\frac{\hbar\omega_E}{k_BT}\right)}{\left(\exp\left(\frac{\hbar\omega_E}{k_BT}\right)-1\right)^2}\left[1-\frac{\partial\ln\omega_E}{\partial\ln T}\right] \tag{7}$$

where R is the gas constant. The last term is absent for the upper temperature-independent frequency, ω_{E2}. The results of the fit are shown in Fig. 1. The change of the specific heat at T_p is reproduced by assuming the jump of the lower Einstein frequency from $\hbar\omega_{E1}$=27.4K to $\hbar\omega_{E1}$=30K at this temperature with ω_{E2} being constant. Above T_p the lower frequency mode is temperature dependent and the overall behavior of $\omega_{E1}(T)$ is shown in Fig.2. Note that the strong temperature dependence of the lowest rattling phonon mode frequency has been recently observed by the Raman scattering[20,21].

In addition to the anharmonic temperature dependence of the rattling phonon mode which modifies the usual approximation for the Eliashberg function, the conduction electrons couple to the rattling phonons, further renormalizing its energy position and introducing an extra damping. In particular, the spectral function of the low-energy rattling phonon frequency is given by

$$\alpha^2F_{E1}(\omega) = -\frac{\alpha^2(T)}{\pi}\mathrm{Im}D(\omega) = \frac{\alpha^2(T)}{\pi}\frac{4\omega_{E1}(T)\Gamma_0\omega}{\left(\omega^2-\omega_r^2\right)^2+4\Gamma_0^2\omega^2} \quad , \tag{8}$$

where Γ_0 is a anharmonic phonon damping rate. The real part of the rattling phonon self-energy leads to a renormalized phonon frequency,

$$\omega_r^2(T) = \omega_{E1}^2(T) + 2\omega_{E1}(T)\mathrm{Re}\Pi(\omega) \quad . \tag{9}$$

Following previous estimation[14] we have used -Re$\Pi(\omega)$ = 1 meV and $\Gamma_0 = 0.25$ meV. It is important to remember that the electron-phonon coupling parameter $\alpha^2(T) \sim c/\omega_{E1}(T)$ with $c = 1.23\mathrm{meV}^2$ is also temperature dependent.

In Fig.3 we show the Eliashberg function $\alpha_i^2F_i(\Omega)$ including all three contributions for T= 0.5K *i.e.* well below T_p. In contrast to the usual Eliashberg theory the $\alpha^2F(\omega)$ spectrum and the coupling constant $\lambda = 2\int_{-\infty}^{+\infty} d\Omega\alpha^2F(\Omega)/\Omega$ are now temperature dependent. Setting $\mu^* = 0.091$ and solving the Eqs. (1)-(3) in the linearized limit we find the superconducting transition temperature T_c=9.6K. In Fig. 4 we show the results of the solution of the Eliashberg equations for $T = 0.5$K. One finds the typical behavior of a strong-coupling superconductor. In particular, Re$\Delta(\omega)$ shows three peaks at the energy of the rattling phonons and at ω_D. For larger energies it becomes negative reflecting the effective repulsion for $\omega > \omega_D$. Due to presence of the low-energy rattling phonons one obtains a strong renormalization of the quasiparticle mass and we also find $2\Delta_0/k_BT_c \approx 5.0$. The latter is in good agreement with experimentally observed value[12].

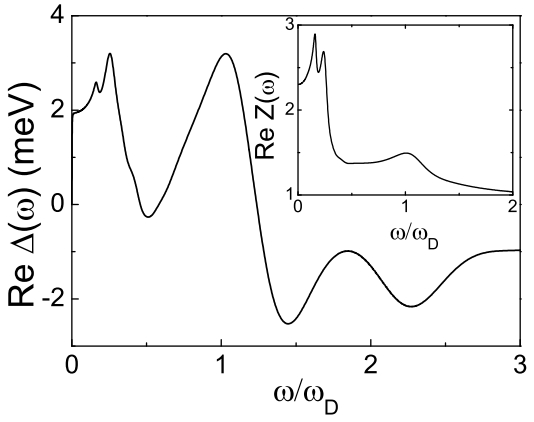

FIG. 4: Calculated frequency dependence of the real part of the superconducting gap function $\Delta(\omega)$ for KOs_2O_6 compounds. The inset shows the renormalization function, $Z(\omega)$ at $T = 0.5$K. Note that we set the cutoff frequency to $8\omega_D$ and introduce a finite damping $\Gamma_0 = 0.25$ meV.

One of the important consequences of the temperature dependent phonon spectrum is slightly anomalous behavior of the superconducting gap as a function of temperature. In Fig.5 we show the temperature dependence of the superconducting gap. One finds a kink at T=T_p due a discontinuous change of the phonon frequency and the corresponding coupling constant. In particular, due to the slight increase of ω_{E1} below T_p the electron-phonon coupling constant decreases which yields lowering of the superconducting gap with respect to its value without the structural phase transition. At the same time, we find overall relatively small modification of the superconducting properties due to structural phase transition. This is consistent with experimental observation[8] which show weak connection between the structural transition and superconductivity in KOs_2O_6. The most important result is that due to temperature dependent low-energy mode the superconductivity is enhanced in KOs_2O_6 compared to its Rb and Cs counterparts. We also find that the electron-phonon coupling strength changes from $\lambda(T > T_p) = 1.7$ towards $\lambda(T < T_p) = 1.6$ and is somewhat lower than estimated previously[8]. This is because a simple application of the McMillan-Dynes formula for estimating λ is probably questionable in KOs_2O_6 due to temperature dependence rattling mode which does not exist in simple Eliashberg theory. Our obtained results although showing an anomaly at T_p cannot explain fully the photoemission data of Ref.[12] as clearly visible from the Fig.5.

Our calculation shows that the superconducting properties of KOs_2O_6 should be sensitive to the external pressure. Due to proximity to the first-order phase transition the application of the pressure may result in a modification of the electron-phonon coupling strength. Such an unusual behavior has been indeed recently found experimentally[23] although the detailed understanding both theoretically and experimentally is still required.

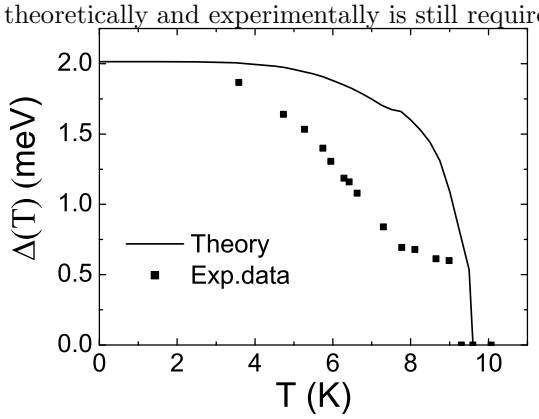

FIG. 5: Calculated temperature dependence of the superconducting gap $\Delta_0(T)$ as determined from $\mathrm{Re}\Delta(T,\omega) = \omega$ for KOs_2O_6. The black squares show the experimental data from Ref. 12 as obtained from the fit of the experimental curve by the Dynes formula.

In summary, we have investigated the superconducting properties of β-pyrochlore KOs_2O_6 compound based on the strong-coupling Eliashberg approach. Analyzing the specific heat data we find that rattling phonon frequency changes discontinuously at the critical temperature of the first order phase transition. Solving the strong-coupling Eliashberg equations with temperature dependent $\alpha^2F(\omega)$, we discuss the consequence of this first order phase transition for the anomalous temperature dependence of the superconducting gap. In particular, we have found that the superconducting gap as a function temperature is anomalous reflecting the temperature dependence of the Eliashberg function.

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